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LINE BROADENING AND SHIFTS OF RARE-EARTH ION ENERGY LEVELS IN S--ETC(10)

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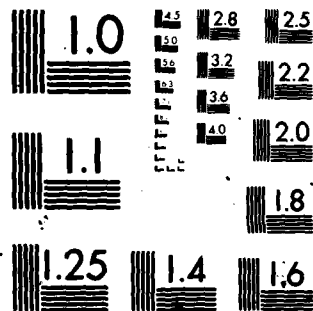
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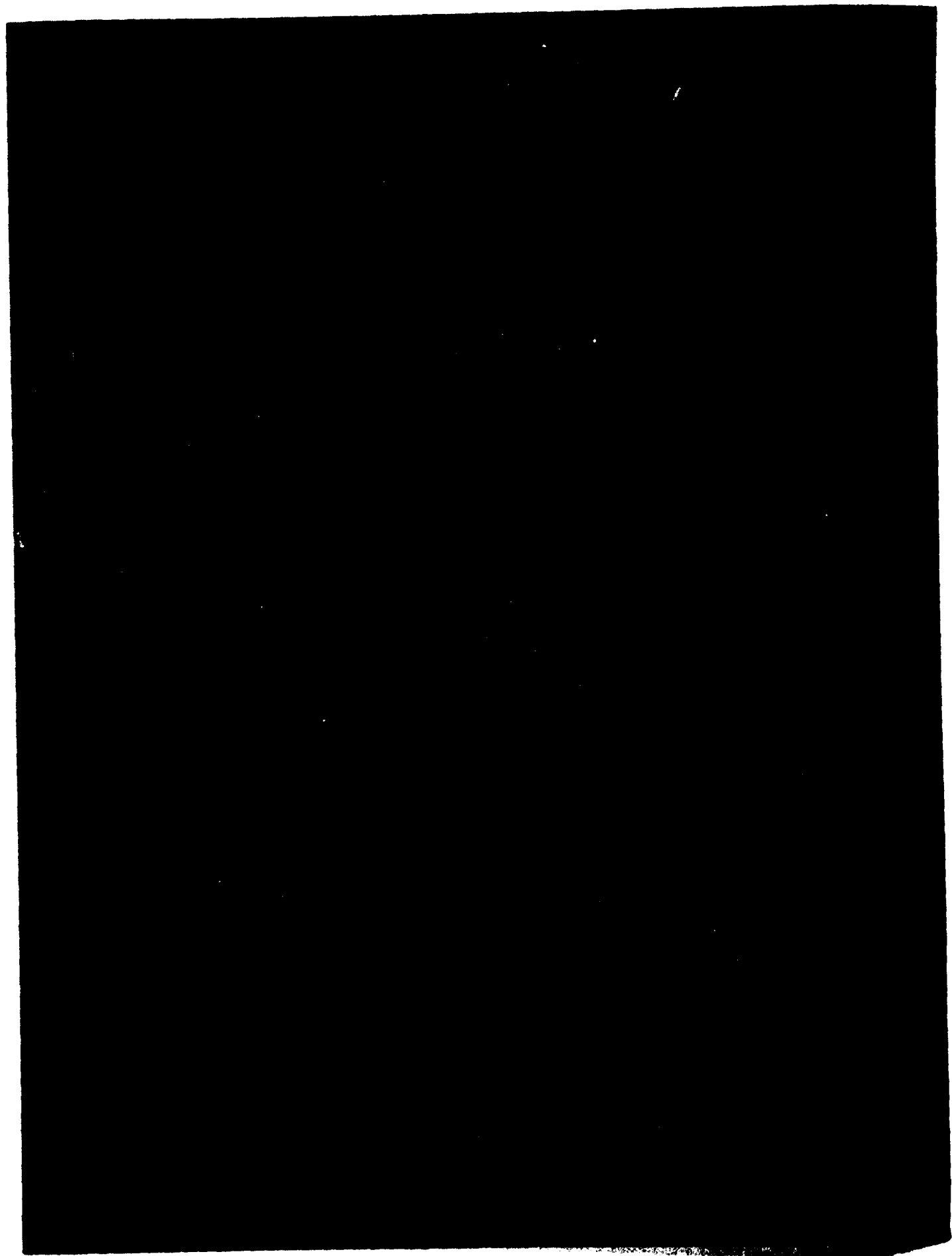
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The usual procedure for calculating inhomogeneously broadened line shapes of rare-earth ions in crystalline solids is to assume a particular form of the interaction Hamiltonian and to proceed by using perturbation theory.<sup>1</sup> The procedure outlined here, although a form of perturbation theory, is presented in a form that will allow for a simpler method of ascertaining the magnitude of various forms of interactions. This method is particularly applicable if computational facilities are available to calculate the crystal-field energy levels of rare-earth ions in various host crystals.

The analysis of rare-earth ions in solids assumes<sup>2</sup> that the Hamiltonian is composed of two parts,  $H_0$  and  $H_1$ . The first part,  $H_0$ , contains the "free ion" parameters; these are chosen so that in the absence of the crystal field an adequate representation of the centroids of the various multiplets is obtained. The second part,  $H_1$ , is the Hamiltonian for the crystal field, written

$$H_1 = \sum_{kq} B_{kq}^+ C_{kq} \quad (1)$$

where the crystal-field parameters,  $B_{kq}$ , are such that  $k$  is even and  $-k \leq q \leq k$ . A sum over the electrons of the configuration  $4f^N$  is implied in equation (1) but will be suppressed, except when necessary for clarity. The  $B_{kq}$  represent the effect of the surrounding host on the rare-earth ion. It is generally assumed that  $H_0$  is not sensitive to local variations within a crystal and that the individual Stark-split rare-earth energy levels are most sensitive to changes in the  $B_{kq}$ . We shall assume so here.

For a small change in the  $B_{kq}$ , we can then write for the shifted energy level

<sup>1</sup>B. DiBartolo, *Optical Interactions in Solids*, John Wiley and Sons, Inc., New York (1968).

<sup>2</sup>B. G. Wybourne, *Spectroscopic Properties of Rare Earths*, Wiley-Interscience, New York (1965).

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$$E_n = E_n^0 + \sum_{kq} \left( \frac{\partial E_n}{\partial B_{kq}} \right)^0 \delta B_{kq} , \quad (2)$$

where  $H|n\rangle = E_n^0|n\rangle$ ,  $H = H_0 + H_1$ , and the superscript indicates the evaluation of the quantities in the absence of any change in  $B_{kq}$ . The change in the energy separation between two energy levels  $n$  and  $m$  due to a change in the  $B_{kq}$  can be written

$$\omega_{nm} - \omega_{nm}^0 = \sum_{kq} f_{kq}^{nm} \delta B_{kq} , \quad (3)$$

where

$$\begin{aligned} \omega_{nm} &= E_n - E_m, \\ \omega_{nm}^0 &= E_n^0 - E_m^0, \text{ and} \\ f_{kq}^{nm} &= \left( \frac{\partial E_n}{\partial B_{kq}} - \frac{\partial E_m}{\partial B_{kq}} \right)^0 . \end{aligned}$$

Now if we perform an average appropriate for a particular physical model of the  $\delta B_{kq}$ , we obtain for the line shift

$$\Delta_{nm} \equiv \overline{\omega_{nm}} - \omega_{nm}^0 = \sum_{kq} f_{kq}^{nm} \overline{\delta B_{kq}} , \quad (4)$$

where the  $f_{kq}^{nm}$  are independent of the particular ensemble of  $\delta B_{kq}$ . Similarly, averaging the square of equation (3) results in

$$\overline{(\omega_{nm} - \omega_{nm}^0)^2} = \sum_{kq, k'q'} f_{kq}^{nm} f_{k'q'}^{nm} \overline{\delta B_{kq} \delta B_{k'q'}} . \quad (5)$$

Squaring the result in (4), subtracting from (5), and reducing the resulting expression, we have

$$\Gamma_{nm}^2 = \sum_{\substack{kq \\ k'q'}} f_{kq}^{nm} f_{k'q'}^{nm} \left( \overline{\delta B_{kq} \delta B_{k'q'}} - \overline{\delta B_{kq}} \overline{\delta B_{k'q'}} \right), \quad (6)$$

where  $\Gamma_{nm}^2 = \overline{\omega_{nm}^2} - (\overline{\omega_{nm}})^2$ . The quantity  $\Gamma_{nm}^2$  is the second moment of the distribution of shifts and can be a useful measure of the line width. Higher moments of the distribution may be calculated via the above procedure.

The result given in equation (6) for the line broadening is very general at this point. The resultant broadening theory can be applied to impurities, concentration effects, lattice vibrations, and a variety of other effects. The particular type of broadening determines the origin of the  $\delta B_{kq}$  and the type of averaging to be considered appropriate for that particular mechanism. The derivatives  $\partial E_n / \partial B_{kq}$  which are common to any mechanism can be readily calculated by using Feynman's theorem,<sup>3</sup> together with the wavefunction characterizing the level  $E_n$ , so that the calculation of the  $f_{kq}^{nm}$  is straightforward.

As a specific example of the use of the results given in equations (4) and (6), we consider an impurity rare-earth ion (or one of the constituents) in a solid in which a rare-earth ion is a stoichiometric constituent. Consider an electron at  $\vec{r}$  on the impurity ion at the origin and an electron at  $\vec{r}_1$  from the nucleus of a host rare-earth ion at  $\vec{R}_p$ . The interaction of these two electrons can be written<sup>4</sup>

$$H' = \frac{e^2}{|\vec{R}_p - \vec{r} + \vec{r}_1|} = e^2 \sum_{\substack{kq \\ aq}} (-1)^a \left[ \begin{matrix} 2k+2a \\ 2k \end{matrix} \right]^{1/2} \langle k(q) a(a) | k+a(q+a) \rangle \\ \times r^k C_{kq}(\hat{r}) r_1^a C_{aq}(\hat{r}_1) \frac{C_{k+a, q+a}^+(\hat{R}_p)}{R_p^{k+a+1}}, \quad (7)$$

<sup>3</sup>E. Merzbacher, *Quantum Mechanics*, John Wiley and Sons, Inc., New York (1961), p 396.

<sup>4</sup>B. R. Judd, *Angular Momentum Theory for Diatomic Molecules*, Academic Press, New York (1975), p 98.



where  $\binom{2k+2a}{2k}$  is a binomial coefficient,  $\langle k(q)a(\alpha) | k+a(q+\alpha) \rangle$  is a Clebsch-Gordon coefficient and, as in equation (1),  $-a \leq \alpha \leq a$ . Also, as in equation (1) the sum over the electrons of both rare-earth ions is assumed. The result given in equation (7) can be written

$$H' = \sum_{kq} r^k \delta A_{kq}^+ C_{kq} \quad , \quad (8)$$

so that the total  $\delta A_{kq}$  due to all the ions is, summing over host sites  $p$ ,

$$\delta A_{kq} = e^2 \sum_{\alpha\alpha} (-1)^\alpha \left[ \binom{k+a+q+\alpha}{k+q} \binom{k+a-q-\alpha}{k-q} \right]^{1/2} \frac{r_1^a C_{\alpha\alpha}^+(\hat{r}_1) C_{k+a, q+\alpha}(\hat{R}_p)}{R_p^{k+a+1}} \quad , \quad (9)$$

where we have used the fact that the Clebsch-Gordon coefficient in equation (7) can be written in terms of binomial coefficients.<sup>5</sup> If we further assume, as in the point charge model, that the  $B_{kq}$  are related to the  $A_{kq}$ , we can write  $\delta B_{kq} = \langle r^k \rangle \delta A_{kq}$  where the  $\langle r^k \rangle$  contain the shielding factors and other quantities used in the point charge model.

The rare-earth ions of the host material are in particular energy levels, so that we can replace  $r_1^a C_{\alpha\alpha}^+(\hat{r}_1)$  by its expectation value as

$$Q_{\alpha\alpha}(j_p) = \langle j | r_1^a C_{\alpha\alpha}^+(\hat{r}_1) | j \rangle \quad , \quad (10)$$

where the subscript  $p$  on  $j$  indicates the rare-earth ion at  $\vec{R}_p$ . The quantity  $-eQ_{\alpha\alpha}(j)$  is the multipolar moment of the host rare-earth ion in the electronic state  $j$ . Let  $\overline{Q_{\alpha\alpha}}$  be the thermal average of the multipolar moment at site  $p$ ,

$$\overline{Q_{\alpha\alpha}} = \sum_{j_p} \exp(-\beta_0 E_{j_p}) Q_{\alpha\alpha}(j_p) / Z \quad , \quad (11)$$

<sup>5</sup>M. E. Rose, *Elementary Theory of Angular Momentum*, John Wiley and Sons, Inc., New York (1957).

where  $Z = \sum_j \exp(-\beta_0 E_j)$  and  $\beta_0 = 1/kT$ . The average  $\overline{Q_{aa}}$  is independent of  $p$  if we neglect the interaction of the constituent ions. Also in equation (11), degeneracies, if they exist, must be taken into account in both  $\overline{Q_{aa}}$  and  $Z$ . Using in equation (9) the result given in (11), we can write

$$\overline{\delta A_{kq}} = \sum_{a\alpha} G_{qa}^{ka} \overline{Q_{aa}^+} S_{k+a, q+\alpha} \quad (12)$$

where

$$G_{qa}^{ka} = (-1)^a \left[ \begin{pmatrix} k+a+q+\alpha \\ k+q \end{pmatrix} \begin{pmatrix} k+a-q-\alpha \\ k-q \end{pmatrix} \right]^{1/2}$$

and

$$S_{NM} = e^2 \sum_p \frac{C_{NM}(\hat{R}_p)}{R_p^{N+1}}.$$

Since  $\overline{\delta B_{kq}} = \langle r^k \rangle \overline{\delta A_{kq}}$ , we can use equation (12) in (4) to obtain the shift as a function of temperature in the transition  $E_n - E_m$  of the rare-earth ion at the origin. This shift contains a term that persists at absolute zero which arises from the fact that the multipolar contributions have not been contained in the zero-order crystal-field parameters. This shift at absolute zero can be subtracted out so that the relative shift is meaningful.

In obtaining the average of  $\delta A_{kq} \delta A_{k'q'}$ , terms of the form  $Q_{a\alpha}(j_p) Q_{b\beta}(j_{p'})$  arise in the cross products occurring in equation (9). The average values of these products can readily be shown to be

$$\overline{Q_{a\alpha}(j_p) Q_{b\beta}(j_{p'})} = \overline{Q_{a\alpha} Q_{b\beta}} \delta_{pp'} + (1 - \delta_{pp'}) \overline{Q_{a\alpha}} \overline{Q_{b\beta}} \quad (13)$$

where the first term is the thermal average of a product of multipolar moments on the same site. It is assumed that the multipolar moments at different sites are uncorrelated. Using the result (13) we have

$$\overline{\delta A_{kq} \delta A_{k'q'}} - \overline{\delta A_{kq}} \overline{\delta A_{k'q'}} = \sum_{a\alpha} G_{q\alpha}^{ka} G_{q'\beta}^{k'b} \left[ \overline{Q_{a\alpha}^+ Q_{b\beta}^+} - \overline{Q_{a\alpha}^+} \overline{Q_{b\beta}^+} \right] \\ \times T(k+a, q+\alpha; k'+b, q'+\beta)$$

where

$$T(NM; N'M') = e^4 \sum_p \frac{C_{NM}(\hat{R}_p) C_{N'M'}(\hat{R}_p)}{R_p^{N+N'+2}}. \quad (14)$$

The result (14) may then be substituted into (6) to obtain  $\Gamma_{nm}$ . The summations occurring in equations (12) and (14) are similar to those arising in the point charge model; the number of terms is restricted by the point symmetry of the site occupied by the rare-earth ion. The expectation values in equation (10) can be evaluated after the Hamiltonian for the host rare-earth ion has been diagonalized. By the use of Feynman's theorem, the  $Q_{a\alpha}$  are given by

$$\frac{\partial E_j}{\partial B_{a\alpha}} = \langle j | C_{a\alpha}^+ | j \rangle = \frac{Q_{a\alpha}^+(j)}{\langle r^a \rangle}, \quad (15)$$

where the  $B_{a\alpha}$  are the crystal-field parameters for the host rare-earth ion.

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